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Deterioration of Marble
A Retrospective Analysis of Tombstone Measurements in the New York City Area

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DETERIORATION OF MARBLE
A Retrospective Analysis of Tombstone Measurements in the New York City Area

by

R.B. Husar
D.E. Patterson
Center for Air Pollution Impact and Trend Analysis
Washington University
St. Louis, MO 63130

and

N.S. Baer
Conservation Center at the Institute of Fine Arts
New York University
New York, New York 10021

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Project Officer

John W. Spence
Emissions Measurement and Characterization Division
Atmospheric Sciences Research Laboratory
Research Triangle Park, North Carolina 27711

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OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NORTH CAROLINA 27711

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SECTION 1
INTRODUCTION

All materials deteriorate with time due to natural weathering. However, man-induced chemicals in the ambient atmosphere, most noticeably sulfur dioxide, tend to accelerate this weathering process. For chemically reactive materials, including some metals, there is a measurable weathering rate after a few years of exposure to a polluted atmosphere. Other, more weather resistant materials show a measurable deterioration rate only after atmospheric exposures over a decade or century. Marble falls into the latter category, for which reason it has been used extensively for building exteriors and for monuments.

The marble weathering rate is a measurable quantity, but such data do not reveal the cause of the material's loss nor the apportionment of the loss rate between natural and man-induced causes. For this purpose, estimates are necessary of the environmental history of the exposed material. This includes standard meteorological parameters such as rainfall, frost frequency, temperature, etc. More importantly, for the estimation of the man-induced damage, the concentration history of air pollutants is required. However, quantitative estimates of air pollutant concentrations spanning this century are currently not available.

The specific role of each meteorological and pollutant parameter in marble weathering is a formidable task that the National Park Service is studying within the National Acid Precipitation Assessment Program. The purpose of this investigation is to estimate the ambient concentration trends of SO_2 for two sites in the greater New York City area, one with high and another with low SO_2 concentration history, and to compare the marble loss rate at the two sites. Choosing geographically close locations effectively

eliminates the natural weathering as a significant variable, as well as regional scale air pollution influence. Hence, in this case the differences in the weathering rates are reasonably attributable to the man-induced local air pollution. The marble loss rates and the estimated SO_2 concentration histories at the two sites permit an estimate of the damage function, i.e. loss rate per SO_2 concentration.

SECTION 2
TOMBSTONE DATA BASE

New York University, through a program with Task Group G, Effects on Materials and Cultural Resources, within the National Acid Precipitation Assessment Program has compiled measurements of thickness and depth of emblem inscription of marble tombstones in over 23 U.S. Veterans Administration (VA) cemeteries (1). The marble tombstones used in these cemeteries provide a standardized material, i.e., of uniform composition and prepared under controlled conditions, which are useful for studying stone deterioration over the last century as a function of geographic location. Excellent records on individual tombstones are maintained by the Department of Memorial Affairs of the Veterans Administration. In this study, measurements of tombstones recorded by New York University for Cypress Hills and Long Island cemetaries were analyzed. Furthermore, only tombstones prepared from Vermont marble, which is a fine grain stone, were analyzed.

Weathering of most tombstones is most pronounced at the top. This is most apparent from the rounded tombstone edges and from decreasing depth of inscriptions. The part of the stone which is in the ground usually exhibits very little weathering. In most cases, the surface near the ground still carries its original smoothness. The detailed physical chemical mechanisms for this preferential weathering at the top are not clearly documented and are not part of the investigation.

In this investigation, the marble deterioration rate is obtained from the difference between the bottom and the top tombstone thicknesses. The tombstone measurements for the Cypress Hills cemetary are presented in Fig. 1, which shows the bottom-top difference versus inscription date. The earliest tombstones were from 1923, the latest from 1980. The most significant

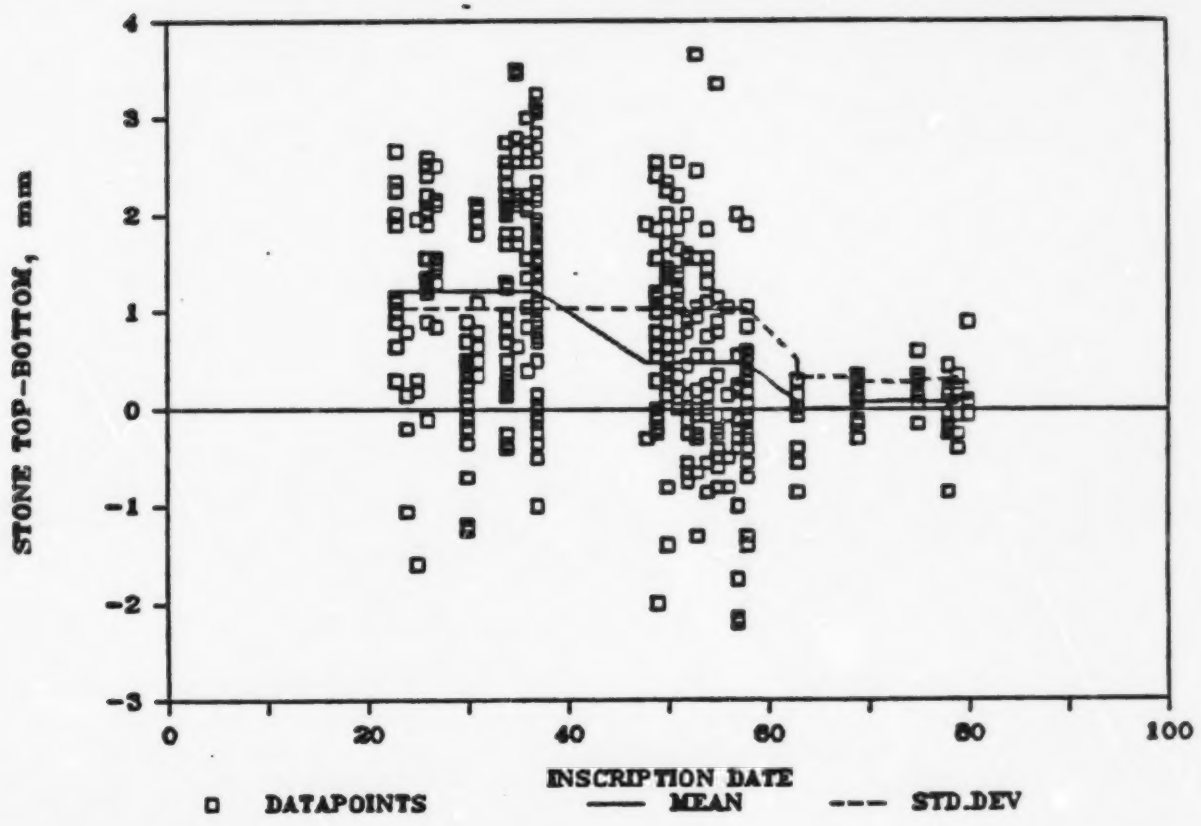

Fig. 1. Marble tombstone loss of thickness, computed as (bottom-top) difference, versus year of tombstone placement at Cypress Hills cemetery, Brooklyn, N.Y. The mean and standard deviation for thickness loss for each placement year cohort are indicated as solid lines.

feature of the data set is the significant scatter of marble loss for each inscription year. In fact, the standard deviation of the bottom-top difference is comparable to the deviation of the mean from zero; i.e., the noise is comparable to the signal.

It is instructive to explore further the nature and possible origin of the noise. Fig. 2 depicts the temporal trend of the left-right difference of the stone thickness. Fig. 2 reflects the fact that the standard deviation of left-right differences is approximately 1 mm, and that its mean is essentially zero for all ages. The orientation of the Cypress Hills tombstones was between 150 and 170 degrees from north, which suggests that preferential weathering is not a significant source for this difference. This suggests that the left-right difference is primarily a measure of the manufacturing tolerance for the tombstones. It is apparent from Fig. 2 that such manufacturing variability is random with respect to left and right. More importantly, the magnitude of the observed left-right variation is comparable to the magnitude of the variation in the bottom-top difference. Consequently, the large variability of the bottom-top signal can be meaningfully reduced by averaging over a large number of observations.

The tombstone data shown in Fig. 1 were averaged for three time periods: 1923-1937, 1948-1958, and 1963-1980. The mean values of bottom-top difference are also displayed in Fig. 1. For the tombstones exposed since the 1920s, the average loss was 1.2 mm while those exposed since the 1950s had losses of 0.7 mm. The few data points for the 1970s show negligible bottom-top difference.

The comparable tombstone loss data set for the Long Island cemetary is shown in Fig. 3. Here again, the large scatter is the dominant feature, the noise being comparable to the signal. However, there is evidence of decreasing

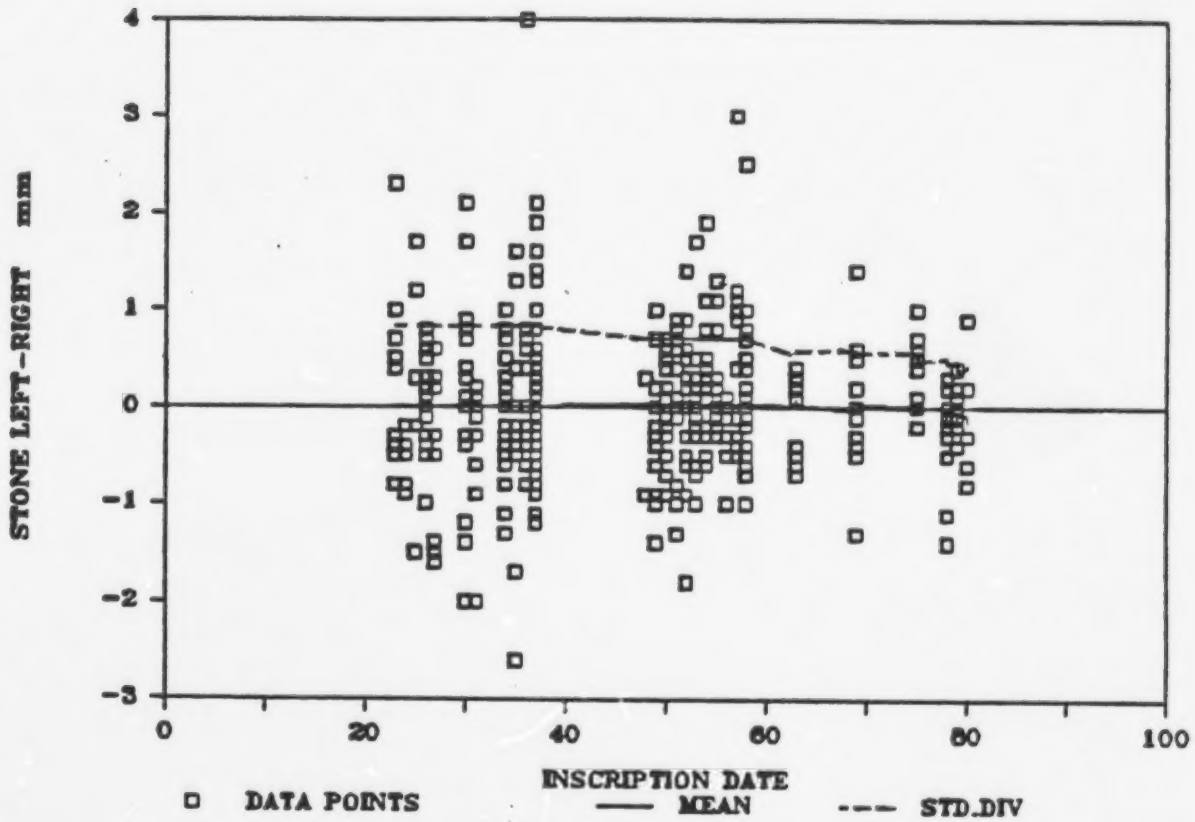

Fig. 2. Left-right differences in thickness at top of tombstones at Cypress Hills cemetery. The mean (typically zero) and standard deviation are indicated as lines.

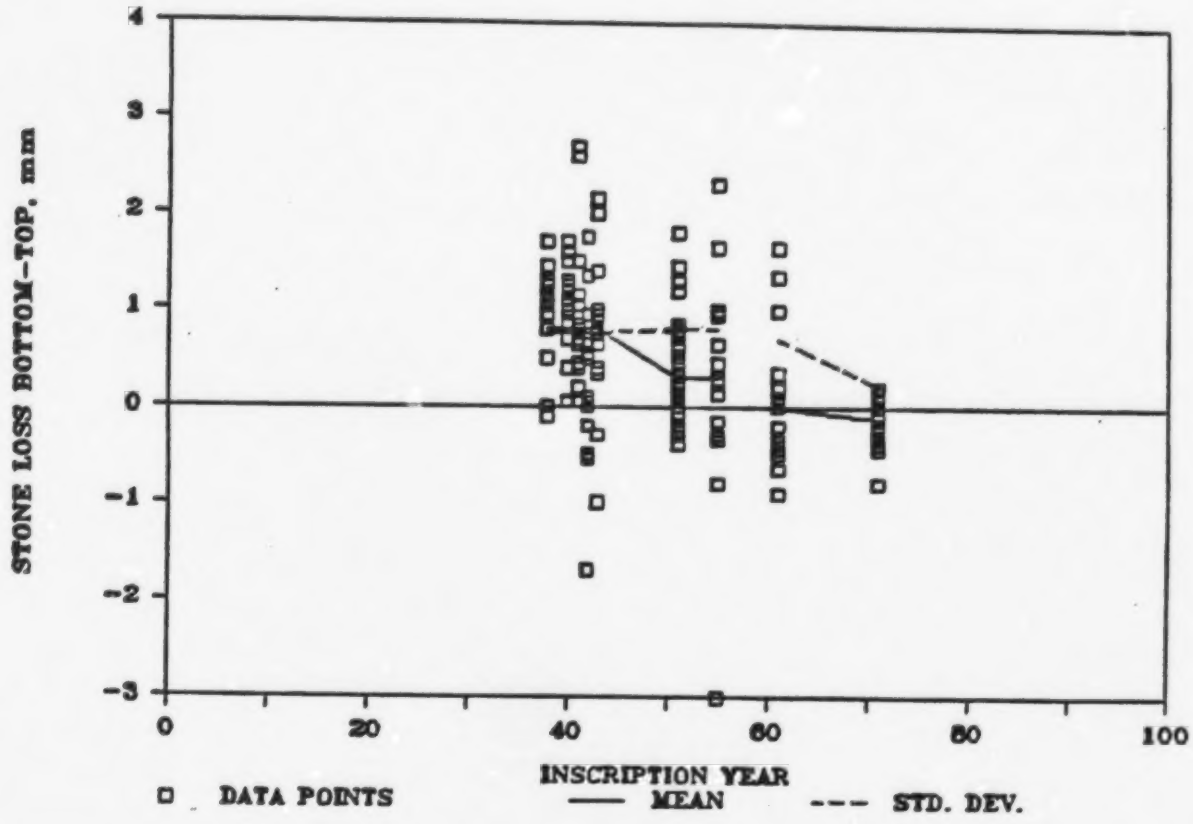

Fig. 3. Marble tombstone loss of thickness, computed as (bottom-top) difference, versus year of tombstone placement at Long Island cemetery, N.Y. The mean and standard deviation for thickness loss for each placement year cohort are indicated as solid lines.

loss for younger stones. The corresponding means indicate that the stones from the 1940s had about 0.8 mm loss, compared to essentially zero mean loss for the recently exposed stones.

The results of the Cypress Hills and Long Island tombstone data are compared in Fig. 4 as weathering rates, i.e., the rate of loss of marble surface in mm/100 years. The rates were derived by choosing representative years with large sample sizes and computing mean loss in mm for each cohort group. The increase in marble loss between successive representative years then yields the loss rate representative for the intervening range of years. For the 1930-55 time period, the Cypress Hills site shows a weathering rate of about 1.5 mm per century, while the Long Island site is about half of that. For the period 1955-80, the corresponding weathering rates were about a factor of two lower for both sites.

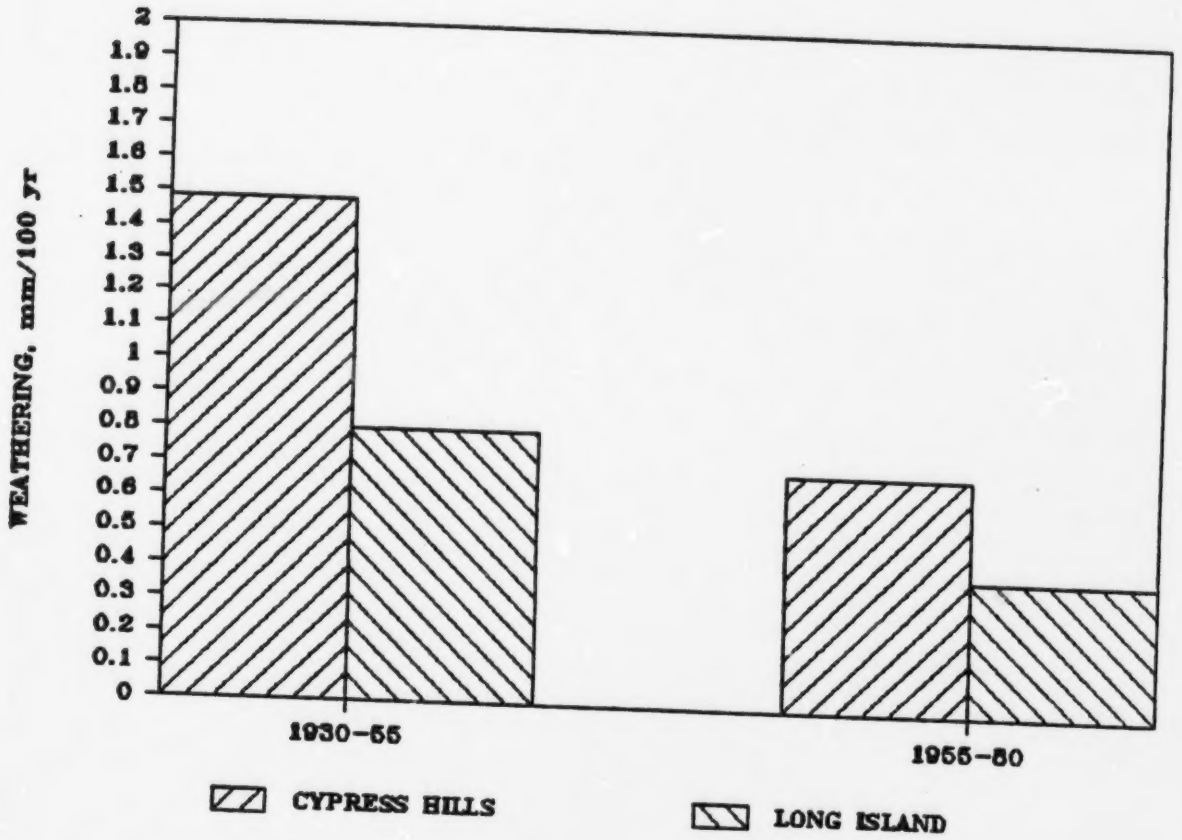

Fig. 4. Comparison of estimated marble tombstone surface loss rates over years 1930-1955 and 1955-1980 at Cypress Hills and Long Island cemeteries.

SECTION 3
AMBIENT CONCENTRATIONS OF SO_2

An estimate of the ambient concentrations of SO_2 at the cemeteries between 1930 and 1980 is now needed in order to derive the weathering rate of Vermont marble in terms of SO_2 concentration. In another study, SO_2 ambient concentrations were estimated using regional and local modeling techniques for New York City, specifically Manhattan's Central Park, from 1880-1980 (2). The estimated retrospective SO_2 concentrations for Central Park are shown in Figure 5. Also shown in Figure 5, is a 15-year trend of measured SO_2 concentration for Central Park (3)(4)(5). The SO_2 trend data indicates about 0.12 ppm in 1960, rising to over 0.20 ppm in 1965, followed by a sharp drop in the late 1960's. It is assumed that the ambient concentration of SO_2 continues to decline in the 1970's.

For this analysis, it is necessary to establish the concentration trends for the Cypress Hills and Long Island cemeteries. In order to estimate these trends, it was assumed that the concentration ratio among Central Park and the two cemeteries has remained constant over the past years. The concentration ratio was obtained from SO_2 concentration maps drawn for the years 1969-71, as reported by Eisenbud et al. (5). Maps for January 1969 and July 1970 are reproduced in Fig. 6a, b. It is evident that the highest concentrations have been measured on Manhatten and adjacent sectors of Brooklyn, Queens and the Bronx. This gradient is particularly pronounced for the winter season. Based on these maps, the SO_2 concentration at the Cypress Hills is about 60% ± 20% of the Central Park while the Long Island site is 30% ± 20% of the Central Park SO_2 concentration.

The above analysis of retrospective concentration estimates demands further discussion of uncertainties. The uncertainty of emissions is estimat-

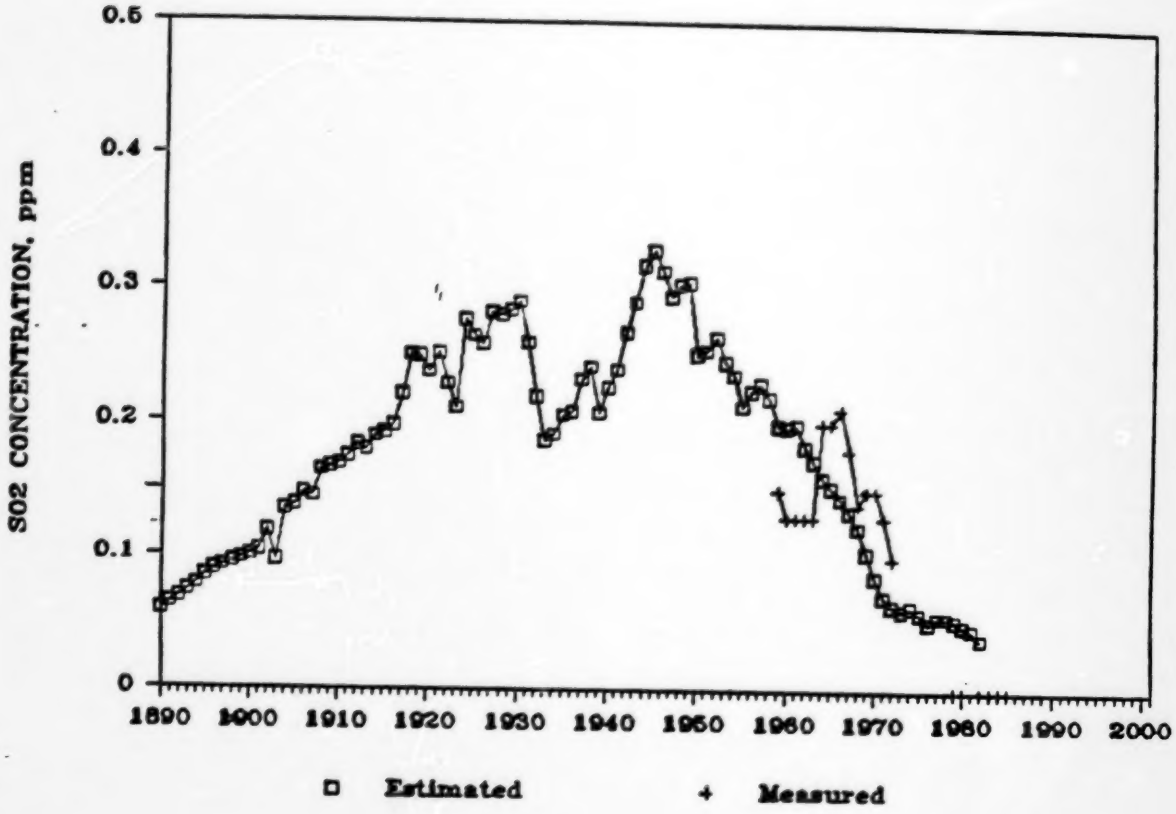

Fig. 5. Retrospective estimate of SO_2 concentration in Manhattan's Central Park 1890-1981.

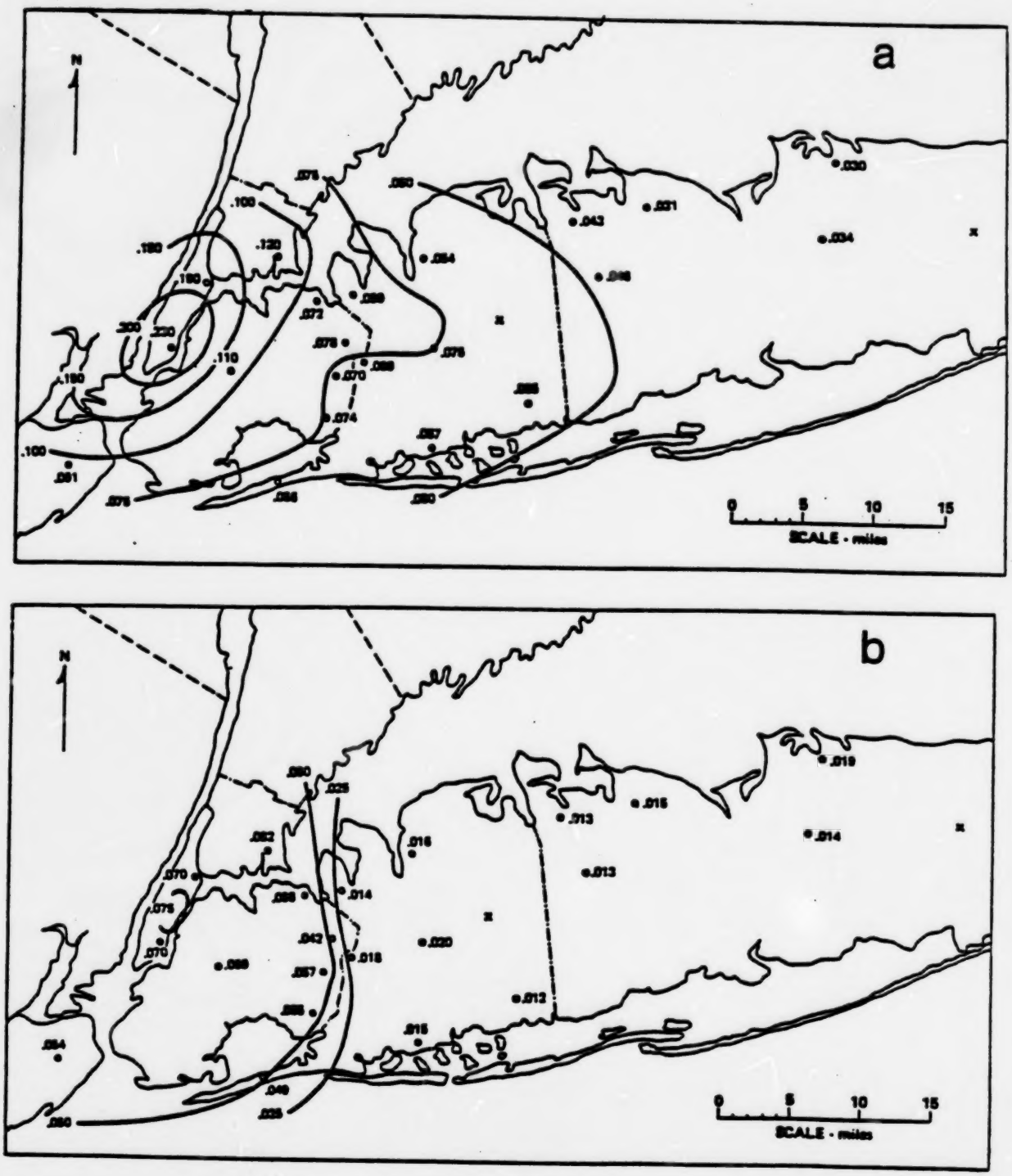

Fig. 6. Observed SO_2 concentration fields for the New York City area, (a) January, 1969; (b) July, 1970.

ed to be about a factor of two around the turn of the century, declining for recent years. It is worth pointing out, however, that for the present consideration it is not vital whether the absolute magnitude has been correctly estimated. The important question is whether the relative variation in time at the two sites is properly assessed. The spatial pattern of emissions in the New York City area has undoubtedly been changing since the turn of the century. It is probable that the emission field in the early decades of the century was much more concentrated around Manhattan while the impact of railroads may have been higher at both sites. Consequently, it is probable that the concentration ratios of Central Park to Cypress Hills and Long Island were actually higher in the early years.

SECTION 4
MARBLE DAMAGE FUNCTION

It is now possible to explore whether the weathering rate of Vermont marble is influenced by the SO_2 concentration. Sulfur oxides are known to promote marble deterioration (6)(7)(8). In a recent field exposure study of marble tiles, the erosion of marble was found to correlate with time of surface wetness; however, the ambient levels of SO_2 were too low to observe a significant effect for the 30-month exposure period (9).

This investigation is based on the assumption that the meteorological conditions for the Cypress Hills and Long Island cemeteries do not differ greatly for the period 1930-80. Since the cemeteries are located less than 40 miles apart, the average meteorological conditions, especially those parameters affecting time of surface wetness, are expected to be quite similar.

A plot of the weathering rate of Vermont marble and corresponding average of SO_2 concentration that were obtained for Cypress Hills and Long Island cemeteries is shown in Fig. 7. Even though there is considerable error associated with each data point, a linear relationship is observed that shows increased weathering with increasing SO_2 concentration. The slope of the linear relationship is 10 mm per century per ppm of SO_2. This value constitutes the best estimate of erosion rate of Vermont marble as a function of SO_2 concentration.

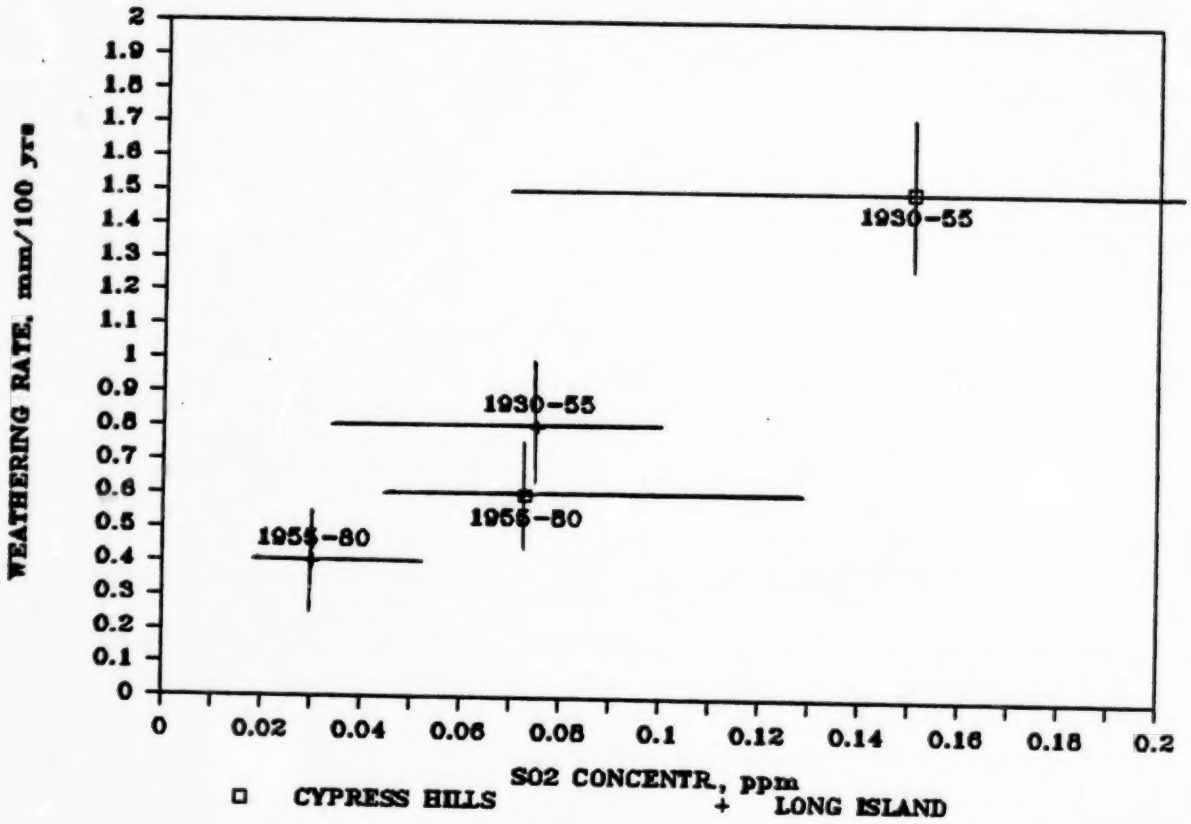

Fig. 7. Preliminary marble SO_2 damage function, relating estimated marble loss rates to estimated average SO_2 concentrations over two time periods at the two cemeteries.

SECTION 5
DISCUSSION AND INTERPRETATION OF RESULTS

The main results of this work are shown graphically in Figure 7, in which the difference in the top-bottom stone width is plotted against the SO_2 concentration. The data points therein may be used to define an empirical damage function, whereby the damage is expressed in units of top-bottom difference as a function of the concentration of the damaging agency, SO_2.

The Vermont marble stone damage function defined in Figure 7 demands examination from at least two distinctly different points of view: a statistical uncertainty of the two physical variables themselves, and secondly the physical-chemical meaning of those variables as indices of the damage and damaging agent, respectively.

The uncertainty of SO_2 concentration estimate for the two sites at each time period has been dealt with in a companion report, "SO_2 Concentration Estimates for New York City, 1880-1980". The resulting uncertainty is depicted in Fig. 7 as horizontal bars indicating the estimated range of SO_2 concentration. It is evident that historical SO_2 reconstruction leaves a great deal of uncertainty in the SO_2 concentration.

The uncertainty bars on the marble weathering rate arise from the averaging over roughly one hundred data points within each time period, and the normality assumption that the standard deviation in the mean decreases as the inverse square root of number of observations. The bars in each case correspond to 2 sigma limits, which are expected to represent approxmately 95% confidence limits for these heavily averaged values. A major reason for averaging over large time periods is to assure acceptable uncertainty in the mean values. For periods of a few years, the confidence limits are comparable to or larger than the mean.

The above discussion states uncertainties associated with the chosen physical parameters. A meaningful interpretation of Figure 7 in terms of a damage function demands the examination of how well the top-bottom difference represents the damage and how well the SO_2 concentration represents the damaging agent.

There are innumerable measures of stone deterioration including chemical parameters such as depth of gypsum layer and physical parameters such as surface roughness and top-bottom width difference. Examination of the different measures led Baer to the top-bottom difference as the most conveniently measured and meaningful loss parameter. It appears indeed to be a reasonable choice, considering the simplicity of its measurement. An inherent assumption in the use of the top-bottom difference is that given a constant physical and chemical environment the top-bottom difference will be linearly increasing with time, i.e. that the loss rate in the first ten years of exposure will be the same as during the second century of exposure, for fixed environmental conditions.

There is empirical evidence that the loss rate of a marble stone in fact is different in the first years of exposure, while the stone retains its polished surface. The phenomenon is illustrated for the tombstone data set from Philadelphia, which has the largest sample size and loss signal. In Figure 8 the top-bottom difference is plotted versus the exposure time, indicating that during the initial 30 years the top and bottom stone widths are comparable. Following the period of constant stone width, there is a monotonic decline with increasing age. Hence the loss kinetics is nonlinear in time and this feature deserves more detailed consideration in future analyses.

A tentative physical-chemical explanation may be given for the observed phenomenon. During the initial years of a freshly exposed $CaCO_3$ block, the polish remains, i.e. there is no physical change in the surface characteristics. However, a layer of $CaSO_4$ develops with increasing depth. In this phase there is little physical loss of stone material, but there is an increase in volume since the density of $CaSO_4$ is much less than that of $CaCO_3$. Once the $CaSO_4$ layer becomes sufficiently deep, its enhanced solubility in water, over that of underlying $CaCO_3$, may lead to loosening of the $CaSO_4$ layer and ultimately to its physical loss due to weathering. In this phase, the thickness of the sulfate layer may remain constant and the reduction in thickness may in fact be proportional to the 'sulfation rate'. This in turn may be proportional to an appropriate measure of the atmospheric sulfur. The verification of this hypothesis will require more detailed physical, chemical and statistical analysis.

Another explanation for this phenomenon is supported by the difference in weathering behavior of marbles of different granular structure. As reported by Baer (1), the weathering rates of fine grain stones are higher than large grain stones. It is therefore theorized that the boundaries between grains of crystalline $CaCO_3$ are more susceptible to weathering, including attack by SO_2, than the grain surfaces of the stone. At some point in time during the weathering process, the erosion of the grain boundary reaches a stage where whole grains are washed away. Since physical measurements are made on grain surfaces of the stones, a decrease in stone thickness, i.e. an increase in erosion rate, would be observed during the stage of granular loss. If this concept is correct, it appears (see Figure 8) that granular loss begins after 20 to 30 years for the Vermont stones located at the Philadelphia Veteran's Administration cemetery.

A major question also arises regarding sulfur dioxide being the active agent rather than wet deposition of sulfur or other chemical compounds that are correlated with SO_2. Even if it can be shown that sulfur dioxide concentration is the relevant parameter, the question arises as to what temporal averaging is pertinent. It is likely that the relevant SO_2 averaging would be weighted according to the vulnerability of the marble surface to the ambient SO_2, e.g. weighting SO_2 by time of wetness. If so, the trends in SO_2 concentration in winter months and during nighttime hours may be more appropriate than the annual average values used in this work. Unfortunately, to do so requires both knowledge of the appropriate weighting function and, more importantly, the ability to estimate the concentration trends on such time scales as seasonal and diurnal. Such detailed reconstruction is beyond the scale of the current study.

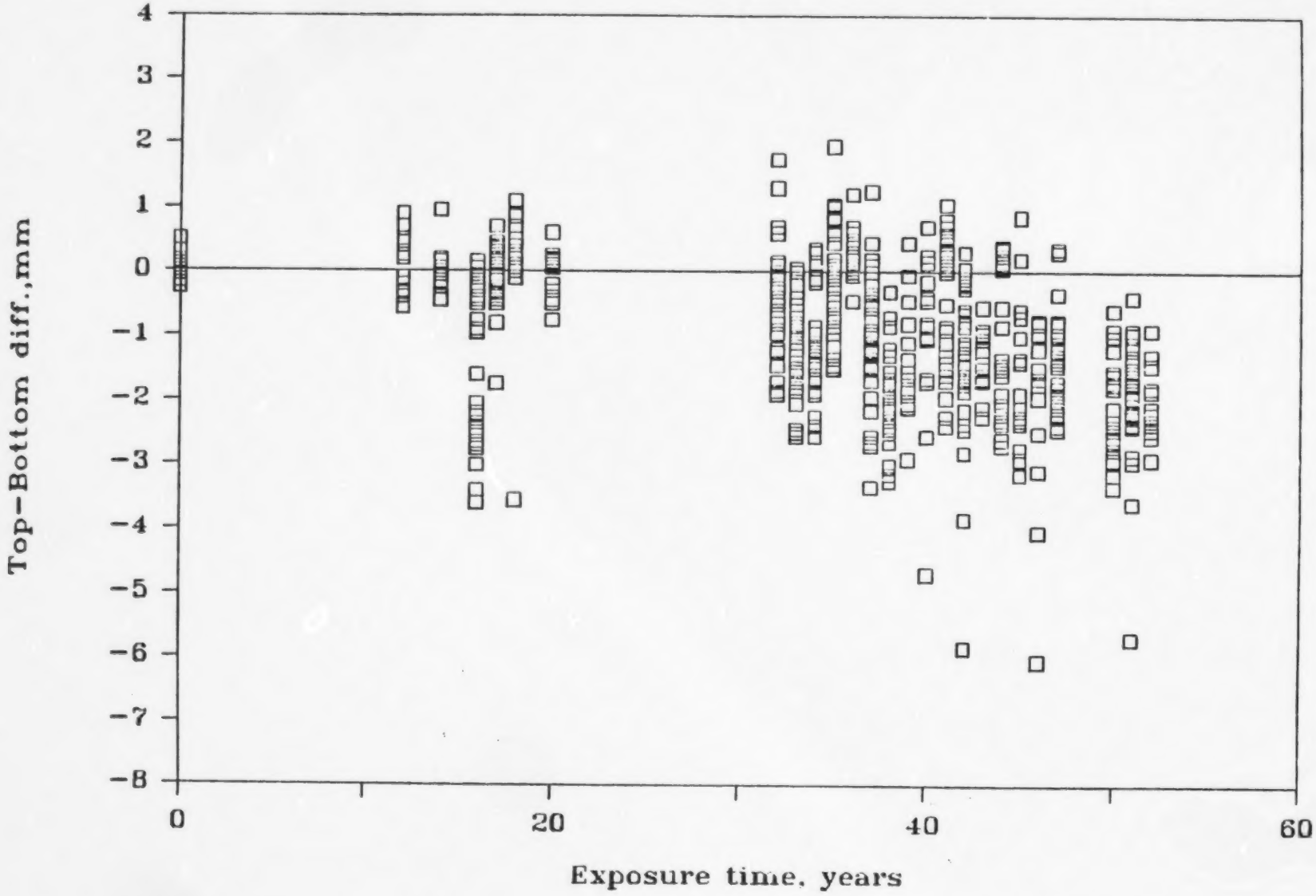

Fig. 8. Marble tombstone loss of thickness versus exposure time for Philadelphia Veterans Administration cemetery.

SECTION 6
CONCLUSION

Other researchers have estimated the weathering rate of marble from the measurements of tombstone. In a very early study, Geikie (10) estimated a weathering rate of 9 mm per century for marble and 0.9 mm per century for granite for Edinburgh cemeteries. In more recent investigations of tombstones, weathering rates of marble were estimated to range from 1 to 2 mm per century (11)(12). Weathering rates derived for tombstones at Cypress Hills and Long Island cemeteries are within the range reported for these recent studies.

Investigations, as described in this report, in which weathering rates of marble were correlated with environmental parameters, are very limited. Meierding, however, found weathering rates of 0.1 mm per century and 1.6 mm per century for marble tombstones in rural U.S. areas where the average rainfall was 20.0 mm and 100 mm, respectively (13).

Based on theoretical considerations, Haynie derived a stoichiometric rate relationship as a function of SO_2 flux, rain (amount and pH), and time of surface wetness for the erosion of marble (9). If one assumes a deposition velocity of 1 cm/sec and a fraction of time when wet of .25 years this relationship predicts an erosion rate of 11 mm per century per SO_2 ppm. The calculated rate represents an upper limit for marble erosion for these conditions.

Based on these recent studies, the value of 10 mm per century per SO_2 ppm that was derived in this investigation appears to be a reasonable estimate of marble deterioration that has occurred in the New York City area.

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